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Blocking phenomena in granular magnetic alloys through magnetization, Hall effect, and magnetoresistance experiments

J. C. Denardin[a)]
Instituto de Física Gleb Wataghin, Universidade Estadual de Campinas, Caixa Postal 6165, Campinas 13.083-970 São Paulo, Brazil

A. B. Pakhomov[b)]
Department of Materials Science and Engineering, University of Washington, Seattle, Washington 98195-2120

A. L. Brandl, L. M. Socolovsky, and M. Knobel
Instituto de Física Gleb Wataghin, Universidade Estadual de Campinas, Caixa Postal 6165, Campinas 13.083-970 São Paulo, Brazil

X. X. Zhang
Physics Department and Institute of Nanoscience and Technology, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong, China

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Magnetization and magnetotransport were measured in Co_xAg_{1-x} granular composites as a function of temperature and applied magnetic field. A transition from blocked to superparamagnetic behavior with increasing temperatures can be observed in magnetization, giant magnetoresistance and the extraordinary Hall effect measurements. However, the blocking temperature determined from magnetotransport measurements is systematically lower than the one estimated from magnetic measurements. This is due to the selective magnetic scattering, which is enhanced for smaller particles, while the magnetization probes the whole particle size distribution. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1542678]

The discovery in 1992 of giant magnetoresistance (GMR) in granular magnetic nanocomposites[1] has generated many studies of both spin-dependent transport and magnetic properties of these materials. Following the study of GMR[1] and extraordinary Hall effect (EHE)[2] in bimetal granular magnetic structures, interesting spin-dependent phenomena in magnetic metal–insulator systems, such as giant Hall effect[3] and large tunneling magnetoresistance,[4] have been reported. EHE, which is believed to be proportional to magnetization,[5] has been used as a powerful tool in studies of different materials and structures, for example thin magnetic metal films[6] or granular systems.[2,3,7–10]

In this letter we show that studies of blocking phenomena and superparamagnetism in magnetic granular alloys with the help of magnetotransport may in some cases give results different from magnetic measurements. Some studies of magnetic relaxation in granular systems by means of EHE[8] and GMR[11] have already been reported. However, magnetotransport is highly sensitive to the details of microstructure, in particular large values of EHE and GMR are associated with the presence of extremely small magnetic particles in the structure.[2,7] If the system has a wide distribution of magnetic particle sizes, smaller particles will give a relatively larger contribution to magnetotransport. Hence, correlation with the overall magnetization of the system, obtained through conventional magnetometry, will be distorted, the difference being larger when the size distribution is wider. We clearly demonstrate this difference in a granular magnetic system (Co_xAg_{1-x} films), by comparing zero-field-cooled (ZFC) and field-cooled (FC) EHE measurements in a small field, EHE thermoremanence and both EHE and GMR hysteresis loops with the corresponding measurements obtained through conventional magnetometry.

The 500-nm-thick granular $Co_x(Ag)_{1-x}$ films with Co volume fraction $x=0.10$, 0.15, and 0.25 were prepared on glass and Kapton substrates in a magnetron cosputtering system, with Co and Ag targets mounted on two separate guns. The Co volume fraction was controlled by the relative sputtering rates, and was then determined by energy-dispersive x-ray spectroscopy using a Philips EDAX XL30 on films deposited in the same run on Kapton. We used the samples deposited on Kapton for magnetic measurements shown in this letter. Structural characterization was performed by transmission electron microscopy (TEM) using a JEOL JEM-3010 ARP microscope, and by x-ray diffractometry. Magnetization and transport properties were measured in a Quantum Design MPMS XL7 system in the temperature range 5–300 K and fields up to 7 T. Resistance and magnetoresistance were measured in bar-shaped samples using the four-probe method. Measurements of Hall resistance were made using the van der Pauw method, without magnetic field reversal.[12]

Figure 1 shows typical x-ray diffraction patterns for $Co_x(Ag)_{1-x}$ samples with (a) $x=0.15$ and (b) $x=0.25$. A sharp peak appears at $2\theta\sim 38°$ that corresponds to the fcc Ag (111) reflection. A second broadened peak at $2\theta\sim 44°$ corresponds to fcc Co (111) phase. No hcp Co was present in the

[a)]Also at: Physics Department and Institute of Nanoscience and Technology, The Hong Kong University of Science and Technology, Clear Water Bay, Kowloon, Hong Kong, China.
[b)]Electronic mail: pakhomov@u.washington.edu

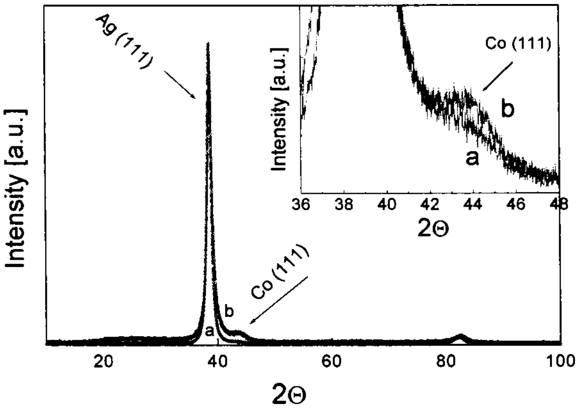

FIG. 1. X-ray diffraction curve for $Co_{0.15}Ag_{0.85}$ sample (a), and $Co_{0.25}Ag_{0.75}$ sample (b). The inset shows details of the (111) Co peak.

samples. The average grain diameters in the sample with $x = 0.25$, evaluated using the Scherrer's formula, are 18.8 nm for Ag and 6.6 nm for Co. The sizes of Ag grains are in good agreement with the average size of microscopic features observed in TEM images. A similar calculation done in the $Co_{0.15}Ag_{0.85}$ sample gave $D \sim 23.9$ nm for Ag. Estimation of the Co size was difficult, because of the poor resolution of the (111) peak (see Fig. 1).

Figures 2(a) and 2(b) show the results of ZFC and FC measurements of both the magnetization M (open symbols) and the Hall resistivity ρ_{xy} (filled symbols), for samples with $x=0.15$ (a) and $x=0.25$ (b), in a field of 200 Oe. Here $\rho_{xy} = (tU_y)/(I_x)$ where t is the thickness of the sample, U_y is the Hall voltage, I_x the longitudinal current. Both the M vs T and ρ_{xy} vs T ZFC curves display maxima, which are not reproduced in the FC measurements. One can associate the positions of the maxima of the ZFC curves with the mean blocking temperature T_b.[13] The extraordinary Hall resistivity is usually assumed to be proportional to magnetization and a

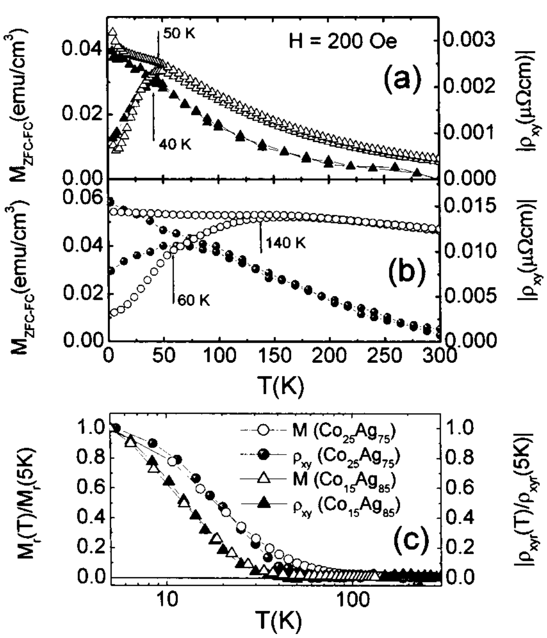

FIG. 2. ZFC and FC magnetization (open symbols) and Hall resistivity (filled symbols) for samples with (a) $x=0.15$ and (b) $x=0.25$, in a field of 200 Oe. (c) Remanent magnetization (open symbols) and remanent Hall resistivity (full symbols) normalized to the values at 5 K, as a function of temperature for $Co_x(Ag)_{1-x}$ samples with $x=0.15$ and $x=0.25$.

TABLE I. The values of the blocking temperature T_b obtained from magnetization (M) and EHE measurements; corresponding calculated values of the mean diameter of magnetic particles D_m and the width of the distribution function (σ_D). D_m determined from x-ray diffraction (XRD) is shown in the last column.

x	T_b (K) M	D_m (nm) M	σ_D M	T_b (K) EHE	D_m (nm) EHE	D_m (nm) XRD
0.10	20	3.8	0.14	20	3.8	⋯
0.15	50	5.2	0.20	40	4.8	⋯
0.25	140	7.2	0.50	60	5.4	6.6

power of resistivity: $\rho_{xys} \propto M_z \rho_{xx}^n$, where n depends on the EHE mechanism.[5] Thus, T_b can be also obtained from the Hall measurements. For the sample with $x=0.1$ (not shown here), both EHE and magnetization curves have the maxima at the same temperature $T_b = 20$ K. However, as seen from the data shown in Fig. 2, no such simple correlations are observed at higher concentrations. The positions of the maxima of EHE are shifted to lower temperature compared to the magnetization curves both for $x=0.15$ [Fig. 2(a)] and $x=0.25$ [Fig. 2(b)], and the difference is larger for the sample with higher Co concentration. The values of T_b from magnetization and EHE measurements are given in Table I. The corresponding mean diameters of the particles (D_m) and the width of the log-normal distribution function (σ_D) of diameter sizes $\{f(D) = 1/\sqrt{2\pi\sigma_D^2 D^2}\exp[-\ln^2(D/\langle D\rangle)/2\sigma_D^2]\}$ were estimated using a model for the relaxation time for a system of superparamagnetic particles[13] and are also shown in the table. The results of D_m and σ_D were also checked through room-temperature measurements of magnetization versus field curves, which were fitted using Langevin functions, properly weighted by the relative contribution of each particle size in a log-normal distribution function.[14] The results of both analysis are very similar, indicating that indeed there is a net increase in the mean grain size and a broadening of the distribution function when the Co volume fraction is increased. The estimated diameters are smaller when determined from the Hall resistance measurements. This result is explained by the correlation between the EHE and particle size distributions.[2,7] While the magnetization is given by the total contribution of the magnetic moment of each magnetic grain, weighted by the corresponding distribution function, the Hall effect is more sensitive to the smaller particles of the system. Figure 2(c) shows the comparison between remanent magnetization and remanent Hall resistivity, for both samples. In this measurement, a field of 7 T was applied at 5 K, then the field was removed and the sample was slowly heated at zero external field. The remanent signals decrease monotonically, and approach zero near the blocking temperature. The logarithmic temperature scale chosen in this figure allows to see the difference in behavior more clearly. The values of the blocking temperatures obtained from these measurements are in excellent agreement with those obtained from the ZFC/FC experiments.

Figure 3 shows magnetization, Hall resistivity and magnetoresistance of sample $Co_{0.25}Ag_{0.85}$ as functions of applied magnetic field at two chosen temperatures: 5 and 50 K. Without taking into account the particle size distribution, one could expect that EHE would scale with magnetization,[5] and GMR with the square of magnetization.[1] However, these re-

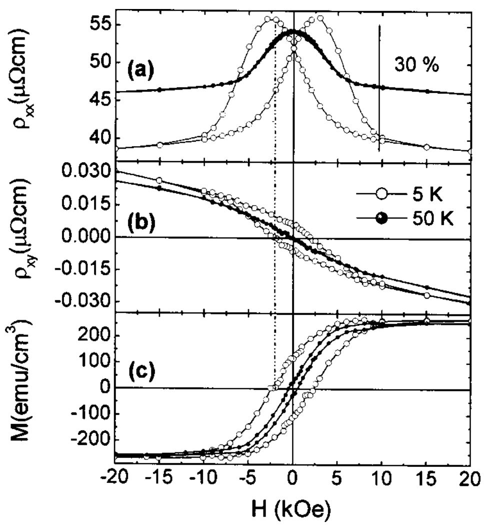

FIG. 3. (a) Magnetoresistance, (b) Hall resistivity, and (c) magnetization for sample with $x=0.25$ as functions of applied field at temperatures of 5 and 50 K.

lationships do not hold at different temperatures. At $T=5$ K, all three curves are hysteretic, with almost the same coercive field. At $T=50$ K, hysteresis is observed only in the magnetization curve, while nonhysteretic behavior is seen in both GMR and EHE. This is consistent with the earlier discussion. At low temperature almost all magnetic particles are blocked, and all three properties respond similarly. From the data one can infer that at 50 K the smallest particles are already superparamagnetic, and such particles play a major role in the magnetotransport properties. On the other hand, the largest magnetic particles are still in the blocked state, resulting in the measured magnetization response with a clear ferromagnet-like character.

In the approximation of large magnetic grains, $r_0 \gg l$, where r_0 is the granular radius and l is the electron mean free path, the EHE coefficient increases with decreasing radius, due to the quasiclassical size effect, as[15] $R_s = R_s^b + 0.2PR_s^s(l/r_0)[1+P(l/r_0)]$, where R_s is the spontaneous Hall coefficient of the granular system, R_s^b is the coefficient for the bulk metal, R_s^s is the contribution from the grain-matrix interface, P is the coefficient of electron reflection from the boundaries. The Green's function calculation for the case of small grains $r_0 \ll l$ also shows that the value of R_s is increasing with decreasing granular size.[16] The fact that the differences in the values of T_b deduced from magnetic and magnetotransport measurements are more apparent at increasing Co volume fractions (Table I) directly reflects the increase in the mean diameter and corresponding width of size distributions. We notice that GMR, as well as EHE, can also be used for probing the spin dynamics in magnetic granular alloys. Indeed, observations of long-term relaxation of magnetoresistance in magnetic granular alloys, which is associated with magnetic relaxation, have been reported in Ref. 11. The difference between GMR and magnetization hysteresis loops observed in this work (Fig. 3) reflects the dependence of spin-dependent scattering on the granule size. However, the sensitivity of measuring GMR in small fields is smaller than that of EHE, because of the quadratic dependence of GMR on magnetization. This is why ZFC and FC measurements of EHE are more informative than ZFC and FC measurements of GMR. It is believed that EHE is proportional to magnetization and to a power of resistivity.[5] Our results indicate that for a superparamagnetic system with a wide distribution of particle sizes this is generally not true. Moreover, as has been shown before[8] one needs to be careful with the correlation with resistivity in these materials, because of coexistence of magnetic and nonmagnetic scattering channels. Theoretically the temperature dependence of ordinary Hall effect in nonmagnetic granular systems in quantum interference regime has been studied in Ref. 17, and that of EHE in granular magnetic systems at high temperatures in Ref. 18. However, there has been no theoretical study of the temperature dependent size effects observed in the present work.

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